

Many-body Floquet theory of laser-induced phase transition in quantum magnets

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We theoretically predict a non-equilibrium phase transition in quantum spin systems induced by laser, which provides a purely quantum-mechanical way for a coherent control of magnetization. Namely, when a circularly polarized laser is applied to a spin system, the magnetic component of laser induces a magnetization normal to the plane of polarization, leading to an ultrafast phase transition. We first demonstrate this numerically for an $S = 1$ antiferromagnetic Heisenberg spin chain, where the topologically-ordered Haldane state gives way to a new state with an emerging magnetization. We then elucidate its physical mechanism by applying a many-body Floquet picture. The theory also indicates that the phenomenon should occur in general quantum spin systems with a magnetic anisotropy. The required laser frequency is in the THz range, and the intensity within a prospective experimental feasibility.

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There is a growing fascination with physics of non-equilibrium systems, which is becoming an important topic in condensed-matter and other fields of physics. For electron systems, a host of novel phenomena induced in non-equilibrium situations, such as photo-induced Mott transitions [1], photo-induced topological transitions [2], etc, have been fathomed. It is a hot topic in cold atoms as well, where quantum simulation is being realized [3]. Now we pose a question: “can we propose novel non-equilibrium phenomena for quantum spin systems?” Spin systems, a basis for the physics of magnets, are many-body in nature and have various applications in engineering and, more recently, in spintronics. This paper proposes a novel, non-equilibrium way to *coherently* control many-body states in *spin systems* with laser. This is in contrast to the control of single quantum states, e.g., qubits, which is becoming important in the field of quantum computation [4]. Instead, we want to develop a method to control the collective phenomena, e.g., phase transitions, with laser.

Interaction between matter and laser, the most powerful tool for coherent control, has been mainly used in electron and atomic systems. So the first question one might ask is “can spin systems be manipulated by laser as well?” The second question, no less important, is “can it be done coherently on a many-body state?” Control of spin systems by lasers is a hot topic due to recent experimental advances [5, 6]. *The key of our study is to use the magnetic component of circularly polarized lasers with photon energy far below the electron energy scale.* It was demonstrated recently that magnetic fields from lasers in the THz regime can directly access to spin dynamics without disturbing the charge degrees of freedom [5]. The merit is that incoherent processes resulting from charge excitations can be prevented. While the setup of Ref. 5 was within the linear response regime, here we extend the avenue to the non-perturbative regime by proposing that

one can realize a “laser-induced phase transition” with perfect quantum coherence. We show that, with circularly polarized laser, we can induce net magnetization in a quantum antiferromagnet. First, it is demonstrated numerically with the infinite time-evolving block decimation (iTEBD) [7, 8] for one-dimensional spin chains. The motion of the induced magnetization can be controlled as desired by shaping the laser pulse, hence provides a new technique for coherent spin manipulation.

The dynamical phase transition requires a theoretical treatment which goes beyond the linear response theory. Floquet theory is becoming a standard theoretical picture for studying quantum systems under periodic driving [2, 9–11]. Namely, the time-periodicity enables us to cast a time dependent problem into a static effective model governed by the Floquet Hamiltonian. This has proved to be a powerful method in the theory of “Floquet topological insulators” [2, 9, 10], and has also been applied to many-body problems such as the photo-induced Mott transition [11]. We usually have to deal with a Hamiltonian in an infinite-dimensional Floquet matrix form as a penalty for going from the time-dependent problem to a time-independent one. We find here, however, a dramatic reduction of the dimension for the Floquet Hamiltonian due to a symmetry by applying the many-body Floquet method to spin systems in circularly polarized laser. The reduced model, which we call the “irreducible Floquet model”, turns out to be a spin system with a static slanted magnetic field. The emergence of the laser-induced magnetization can be understood with this static model, which we confirm by comparing exact diagonalization results with iTEBD. Since the above discussion is applicable also for more than one dimension, the induction of magnetization is independent of the dimension.

While the magnetization can be induced in any dimensions, one-dimensional antiferromagnet has, quan-

tum mechanically, a special interest. The system is known to be gapless if the size of each spin, S , is a half-odd-integer, and gapped if S is an integer. This is the celebrated Haldane's conjecture [12, 13], now established by intensive analytic, numerical, and experimental studies. Specifically, the ground state (GS) of $S = 1$ spin chain, known as the Haldane phase, is topologically protected by a symmetry [14], and is characterized by a "string order parameter." [15] This enables us to demonstrate that the laser-induced non-equilibrium phenomenon we propose here is indeed a phase transition from a state having a topological order into a state with finite magnetization which replaces the topological order. As a further indication that the present transition is genuinely a non-equilibrium process, we show that the necessary strength of the magnetic field can be smaller than the static excitation (Haldane) gap, which is not the case with conventional magnetization processes in static magnetic fields.

Let us begin by introducing the model used in the present study. We consider a one-dimensional Heisenberg antiferromagnet with a Hamiltonian,

$$\mathcal{H}_0 = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2 \quad (J > 0), \quad (1)$$

where the term proportional to D is a magnetic anisotropy of a single-ion type. The model is simple, and also realistic for describing, e.g., organic compounds such as $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2\text{ClO}_4$ (NENP) and $\text{Ni}(\text{C}_5\text{D}_{14}\text{N}_2)_2\text{N}_3(\text{PF}_6)$ (NDMAP). D/J in NENP and NDMAP is estimated from experiments to be about 0.18 [16] and 0.25 [17], respectively. Here we set $D/J = 0.25$.

We consider a sudden switch on of the laser at $t = 0$ starting from the GS of (1) which is in the Haldane phase [18]. The state evolves according to the Hamiltonian,

$$\mathcal{H}(t) = \mathcal{H}_0 - A(e^{-i\Omega t} S_{\text{tot}}^+ + e^{i\Omega t} S_{\text{tot}}^-), \quad (2)$$

for $t > 0$, where $2A$ ($= B$) and Ω are the amplitude and frequency (photon energy) of the laser, respectively, and $S_{\text{tot}}^\pm \equiv \sum_i S_i^\pm$ the raising and lowering operators for the total spin. Here we assume that only the magnetic component of the laser couples to the system, and that the laser is applied from the direction of z -axis. Thus each spin feels a magnetic field rotating in the xy -plane, $B(S_{\text{tot}}^x \cos \Omega t + S_{\text{tot}}^y \sin \Omega t)$, which is expressed above in terms of the spin raising and lowering operators ($S_{\text{tot}}^\pm = S_{\text{tot}}^x \pm i S_{\text{tot}}^y$). The time evolution of the induced magnetization \mathbf{M} ($\equiv \langle \mathbf{S}_i \rangle$ here) calculated with iTEBD is displayed in Fig. 1. The magnetization starts from zero and grows in magnitude with time in a precessional movement. Remarkably, \mathbf{M} tends to point in the z direction, despite the external magnetic field being entirely in the xy -plane.

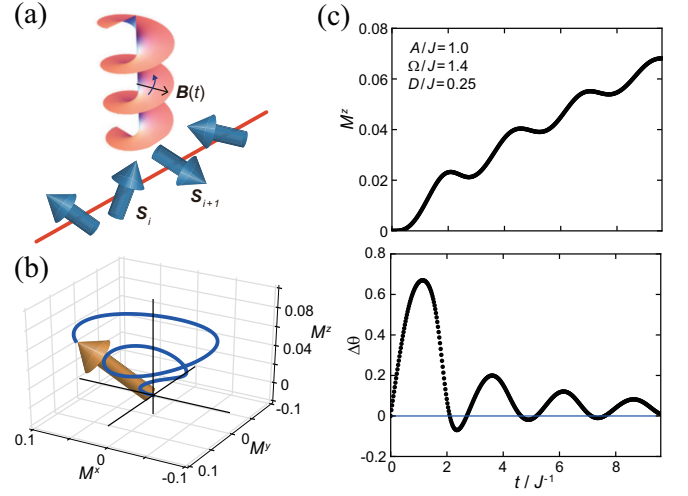


FIG. 1: (color online). (a) A quantum spin system applied by a circularly polarized laser is schematically depicted. (b) A typical evolution of the magnetization induced by the laser, as calculated with iTEBD for $D/J = 0.25$, $A/J = 1.0$, and $\Omega/J = 1.4$. The range of the plot corresponds to about two cycles of the laser (a time interval $0 \leq t \leq 9.6J^{-1}$). (c) Time-evolution of magnetization, M^z , along z -axis, and phase difference, $\Delta\theta$, in the xy -plane between the magnetic field of laser and magnetization.

There are two conditions for the emergence of magnetization. One is the anisotropy of the spin system. In a $\text{SU}(2)$ symmetric system, the spin dynamics becomes trivial since \mathcal{H}_0 commutes with S_{tot}^+ , S_{tot}^- and $S_{\text{tot}}^z \equiv \sum_i S_i^z$. In the present system, the single-ion magnetic anisotropy, i.e., the D term, lifts this constraint. An Ising-type anisotropy (XXZ model) has a similar effect as well. The second condition is to use a *circular* polarization. In linear polarization, emergence of magnetization perpendicular to an applied magnetic field is prohibited by spin-inversion symmetry. Circular polarization breaks the symmetry and induces magnetization initially pointing in the xy -plane, and then slowly moving toward the z -direction. As for the precessional movement, there is an intuitive way to grasp the behavior with a semi-classical spin picture. Fig. 1(c) plots the evolution of M^z , which is non-monotonic. If we compare this with the relative angle between the directions of the external magnetic field and magnetization defined by $\Delta\theta \equiv \Omega t - \arctan(M^y/M^x) \bmod \pi$, the increase and decrease of M^z corresponds to the positive and negative $\Delta\theta$ regions. This indicates that $\dot{\mathbf{M}} \equiv (M^x, M^y, M^z)$ follows a semi-classical equation, $\dot{\mathbf{M}} = \gamma \mathbf{M} \times \mathbf{B}(t)$, where $\mathbf{B}(t) = B(\cos \Omega t, \sin \Omega t, 0)$ is the magnetic field of the laser with γ a positive constant. If we use fields with opposite polarization (left vs. right), the magnetization points in the other direction.

Although we have an intuitive understanding on why the magnetization points in the z -direction, the emergence of the magnetization itself is purely a quantum process. In order to clarify the mechanism, we plot how

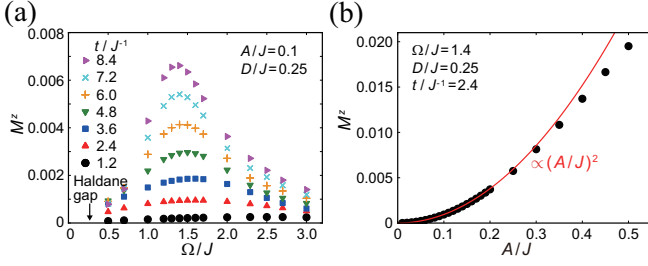


FIG. 2: (color online). (a) Ω -dependence of M^z at various times t . (b) A -dependence of M^z for $t/J^{-1} = 2.4$. The solid line is a fitting to $M^z \propto (A/J)^2$.

the induced magnetization depends on the laser amplitude and frequency in Fig. 2. From the $\propto (A/J)^2$ dependence for small A , we conclude that this is a second-order nonlinear process in terms of the external magnetic field. Figure 2(b) shows the Ω -dependence of M^z at various times t . As time advances, the peak in M^z develops around $\Omega/J \simeq 1.4$, implying a resonance around this frequency. We note that this energy scale is an order of magnitude larger than the Haldane gap $\Delta/J \simeq 0.26J$, which is a hallmark of the contribution from high energy excited states. In the following, we explain the resonance behavior with the help of the Floquet theory combined with exact diagonalization [19].

The Floquet theory is a temporal analogue of the Bloch theorem and maps a system with a time-periodic Hamiltonian $H(t)$ to a static problem. This is done through discrete Fourier transformation of the Hamiltonian $\mathcal{H}(t) = \sum_m e^{-im\Omega t} H_m$, and the time-dependent Schrödinger equation is casted into an eigenvalue equation

$$\sum_m (H_{n-m} - m\Omega\delta_{mn})|\Phi^m\rangle = \epsilon|\Phi^n\rangle, \quad (3)$$

where ϵ is the Floquet quasi-energy level. The solution of the original Schrödinger equation is given as $|\Psi(t)\rangle = \sum_m e^{-i(\epsilon+m\Omega)t}|\Phi^m\rangle$. In many-body Floquet theory, we can think the Fourier index m as a flavor or pseudo-spin which runs from $-\infty$ to $+\infty$ and the term $m\Omega\delta_{mn}$ then acts as an effective “flavor Zeeman term”. Now, the problem becomes time-independent, but a price to pay is that the size of the Hilbert space becomes infinite times larger due to this flavor index. Fortunately enough, however, in spin systems in circularly polarized light, the inter-flavor coupling in the Floquet model is given by

$$H_{+1} = -AS_{\text{tot}}^+, \quad H_{-1} = -AS_{\text{tot}}^-,$$

i.e., the raising (lowering) operator of the flavor index is proportional to the raising (lowering) operator of the total spin, and $[H_0, S_{\text{tot}}^z] = 0$ is satisfied. Therefore, the quantity $(S_{\text{tot}}^z - m)$ is a good quantum number. Thus the Floquet Hamiltonian Eq. (6) becomes block-diagonal in

terms of this quantum number and each block is governed by what we call the “irreducible Floquet Hamiltonian”,

$$H_{\text{Ir}} = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2 - \Omega S_{\text{tot}}^z - B S_{\text{tot}}^x, \quad (4)$$

where $B = 2A$, and the z -direction magnetic field Ω comes from the “flavor Zeeman term”. Different blocks are related through a simple energy shift in an integer multiple of Ω . The obtained effective Hamiltonian, represents nothing but a spin system in a slanted magnetic field $\mathbf{B}_{\text{eff}} = (B, 0, \Omega)$ (Fig. 3(a)). The above discussion is applicable to any lattice structures other than the chains considered here. We can note in passing that the effective description in the Floquet picture has a resemblance with the electron spin resonance (e.g., Ref. 20), where the role of the external magnetic field is now played by the laser frequency Ω . Now it is not difficult to understand the mechanism of the laser-induced magnetization process. Excited states with $S_{\text{tot}}^z = \pm j$ ($j > 0$) are initially degenerate due to the spin inversion symmetry. When the laser is applied, the “longitudinal magnetic field” Ω lifts the degeneracy due to the Zeeman effect and the Floquet energy levels split as $E_j \rightarrow E_j \mp j\Omega$ as shown in Fig. 3(b). The “transverse field” B acts to hybridize the states having different S_{tot}^z ’s. The hybridization becomes larger as the Floquet quasi-energy levels come closer. Since $S_{\text{tot}}^z = +j$ states approaches the GS while $S_{\text{tot}}^z = -j$ states departs from the GS, the GS primarily hybridizes with $S_{\text{tot}}^z = +j$ states, which is precisely why a positive net magnetization appears.

It is non-trivial to take the thermodynamic limit since we have to consider the hybridization with infinitely many levels. To clarify this, we introduce the “magnetic density of states” (MDOS), defined as $\sum_i M_i^z \delta(E_i - E)$, which enables us to capture the behavior of the Zeeman splitting for the entire many-body states. MDOS for various values of Ω calculated with diagonalization of the irreducible Floquet Hamiltonian is shown in Fig. 3(c) for $N = 8$ and $A/J = 0.1$, where it is normalized by 2^N . MDOS, which is zero at $\Omega = 0$ due to the $S_{\text{tot}}^z = \pm j$ degeneracy, is seen to develop positive (negative) peaks, which move to low (high) energies with Ω . When the positive peak overlaps with the GS, the hybridization becomes most prominent, and thus, leads to the resonance-like behavior. The Floquet theory is so powerful that one can reproduce the iTEBD result of the time evolution of M^z . Figure 3(d) plots M^z against Ω at various times calculated by combining the Floquet theory and the exact diagonalization for $N = 8$ [19]. We can see that the result agrees well with the previous one (Fig. 2(b)). The peak is located around $\Omega/J \sim 1.9$, and the deviation from the iTEBD result (peaked around $\Omega/J \sim 1.4$) is attributed to a finite-size effect.

Finally, we show that the current phenomena is actually a laser-induced phase transition from Haldane’s phase to the magnetized phase. To demonstrate this ex-

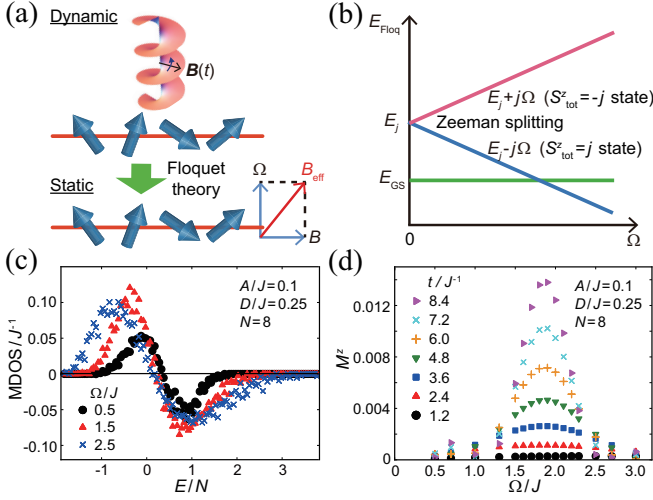


FIG. 3: (color online). (a) The Floquet theory maps the spin system in circularly polarized laser with amplitude B and frequency Ω to a static spin model with a slanted magnetic field $\mathbf{B}_{\text{eff}} = (B, 0, \Omega)$. (b) An effective description of the laser-induced magnetization, where the z -direction magnetic field Ω shifts the many-body energy levels by “Zeeman splitting” of the Floquet energy spectrum. (c) Magnetic density of states (MDOS) in units of states/ J per site for $\Omega/J = 0.5, 1.5$, and 2.5 . (d) Ω -dependence of M^z at various times. Both (c) and (d) are obtained by exact diagonalization of the $N = 8$ irreducible Floquet Hamiltonian Eq. (8).

Explicitly, we calculate the string order correlation [15] with iTEBD. Namely, Haldane’s phase, being a topologically protected phase, has no local order parameter, but is instead characterized by the string order parameter, which is defined as $\lim_{r \rightarrow \infty} \mathcal{O}_{\text{str}}(r)$ from the string-order correlation function, $\mathcal{O}_{\text{str}}(r) = \langle S_0^z \exp(i\pi \sum_{i=0}^{r-1} S_i^z) S_r^z \rangle$ [15]. The string order parameter can capture the Haldane phase, which can be considered as a superposition of states in which an arbitrary number of $|S_i^z = 0\rangle$ ’s are inserted between $|1\rangle$ and $|-1\rangle$ in a Néel order in a S_i^z -basis, e.g., $|\dots, 1, 0, 0, 0, -1, 0, 1, 0, 0, -1, \dots\rangle$. In the initial GS, we are in the Haldane phase with the string order parameter indeed finite as seen in the $t = 0$ result of Fig. 4(a). For $t > 0$, the string order correlation $\mathcal{O}_{\text{str}}(r)$ begins to decay exponentially with distance r . This indicates that the laser-induced phase transition from the Haldane phase to a magnetized phase happens as soon as we have a finite magnetization. Indeed, we find that the decay of the string order has a direct (and even analytic) relation with the uprise of the z -component of magnetization. We can in fact quantitatively correlate the magnetization with the string correlation length ξ (defined by fitting $\mathcal{O}_{\text{str}}(r) \propto \exp(-r/\xi)$). Figure 4(b) plots ξ as a function of M^z . We can deduce an analytic form for the relation between ξ and M^z as follows.

A flip of a single spin from $|0\rangle$ to $|1\rangle$ or from $|-1\rangle$ to $|0\rangle$ in the Haldane phase gives a fac-

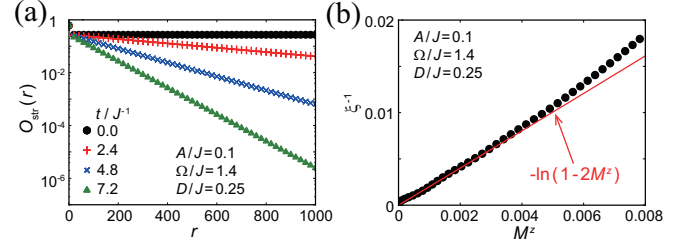


FIG. 4: (color online). (a) The string order correlation function calculated with iTEBD at various times. (b) The inverse of string correlation length, $1/\xi$, plotted as a function of M^z . The result of iTEBD (circles) and the analytical prediction, $\xi^{-1} = -\ln(1 - 2M^z)$ (line) are shown.

tor -1 in the string order correlation function, and acts like a “disorder” in the string-ordered states (e.g., $|\dots, 1, 0, 0, 0, -1, 0, 1, 0, 0, -1, \dots\rangle \rightarrow |\dots, 1, 0, “1”, 0, -1, 0, 1, 0, 0, -1, \dots\rangle$). The probability of having such a disorder is M^z per site, and by estimating the probability of having k disordered sites out of r sites, we are led to an expression,

$$\begin{aligned} \mathcal{O}_{\text{str}}(r) &\simeq S_0 \sum_{k=0}^r (-1)^k {}_r C_k (M^z)^k (1 - M^z)^{r-k} \\ &= S_0 (1 - 2M^z)^r, \end{aligned}$$

where S_0 is the initial (i.e., at $M^z = 0$) value of the string order. Namely, we have $\xi^{-1} = -\ln(1 - 2M^z)$, with which the result in Fig. 4(b) has an excellent agreement in the small M^z region.

Finally let us discuss the experimental feasibility. A pump-probe experiment should be most realistic, where the pump laser is required to be strong and circularly polarized, and the induced magnetization can be measured by Kerr rotation. The necessary pump strength depends on the sensibility of the probe. We make an estimation for NDMAP, whose exchange interaction is $J \simeq 2.8$ meV with $D/J \sim 0.25$ [17]. While we have developed Floquet picture for a continuous laser application, a pulse containing a few cycles of laser can induce a magnetization as illustrated in Fig. 1, and in order to induce $M_z \sim 0.01$ with few cycles we need $A/J \sim 0.1$. This corresponds to $B = 2A = 0.2J \simeq 5.6$ T. The optimum frequency is, from the resonance frequency, $1.4J \simeq 3.9$ meV ~ 1 THz. Currently available intensity of the magnetic field in laser is ~ 0.13 T [5] and is still one order smaller than the required strength. However, THz laser techniques are rapidly advancing and we expect our theoretical proposal become experimentally feasible in the near future.

Finally, let us comment on the outlook of laser-induced magnetization. While we have presented the result for one-dimensional spin systems, our discussions with the Floquet theory do not depend on the dimension nor the size of the spin. There are many interesting spin related phenomena in higher dimensions, such as the effect of

frustration and emergence of spin liquid phase, to name a few. Moreover, quantum spin systems have interdisciplinary spin-offs to e.g., atomic systems in a cavity. It is an interesting future problem to study laser-induced phase transitions in such systems, and the theory presented here is expected to play an important role.

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Supplemental Material

FLOQUET THEORY

Here we elucidate how the application of Floquet theory, which usually leads to an infinite-dimensional matrix equation, to the present quantum spin system can be cast into a finite-dimensional form. Floquet theory is a mathematical technique to treat time-periodic differential equations, which is a temporal analogue of the Bloch theorem for spatially periodic systems. When applied to the time-dependent Schrödinger equation,

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = \mathcal{H}(t)|\Psi(t)\rangle, \quad (5)$$

with time periodicity $\mathcal{H}(t+T) = \mathcal{H}(t)$ (T : period), the Floquet theorem dictates that the solution should have a form $|\Psi(t)\rangle = e^{-i\epsilon t}|\Phi(t)\rangle$, a phase factor involving ϵ called Floquet quasi-energy times a time-periodic wave function (Floquet state) with $|\Phi(t+T)\rangle = |\Phi(t)\rangle$. With both $\mathcal{H}(t)$ and $|\Phi(t)\rangle$ periodic in t , we can make a discrete Fourier transformation,

$$\begin{aligned} \mathcal{H}(t) &= \sum_m e^{-im\Omega t} H_m, \\ |\Phi(t)\rangle &= \sum_m e^{-im\Omega t} |\Phi^m\rangle. \end{aligned}$$

When these are plugged into Eq. (5), the Schrödinger equation is casted into a time-independent eigenequation in a matrix form,

$$\sum_m (H_{n-m} - m\Omega\delta_{mn})|\Phi^m\rangle = \epsilon|\Phi^n\rangle, \quad (6)$$

which can be thought of as an equation for “photon-dressed” (Floquet) modes.

For the present case of one-dimensional Heisenberg antiferromagnet with a single-ion anisotropy in a circularly polarized field with the Hamiltonian

$$\mathcal{H}(t) = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2 - A(e^{-i\Omega t} S_{\text{tot}}^+ + e^{i\Omega t} S_{\text{tot}}^-),$$

the 0th component (H_{n-m} with $n = m$) is $H_0 = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2$, while $H_{\pm 1}$ just corresponds to $H_{\pm 1} = -AS_{\text{tot}}^{\pm}$. The eigenvalue equation (6) then simplifies into a tridiagonal form,

$$\begin{pmatrix} \ddots & & & & & \\ & H_0 - 2\Omega & H_{+1} & 0 & 0 & \\ & H_{-1} & H_0 - \Omega & H_{+1} & 0 & \\ & 0 & H_{-1} & H_0 & H_{+1} & \\ & 0 & 0 & H_{-1} & H_0 + \Omega & H_{+1} \\ & 0 & 0 & 0 & H_{-1} & H_0 + 2\Omega \\ & & & & & \ddots \end{pmatrix} \begin{pmatrix} \vdots \\ |\Phi^2\rangle \\ |\Phi^1\rangle \\ |\Phi^0\rangle \\ |\Phi^{-1}\rangle \\ |\Phi^{-2}\rangle \\ \vdots \end{pmatrix} = \epsilon \begin{pmatrix} \vdots \\ |\Phi^2\rangle \\ |\Phi^1\rangle \\ |\Phi^0\rangle \\ |\Phi^{-1}\rangle \\ |\Phi^{-2}\rangle \\ \vdots \end{pmatrix}, \quad (7)$$

where Ω represents Ω times an identity matrix. The Floquet formalism is schematically depicted in Fig. 5(a), where replicas of the original system are prepared for different photon-dressed (Floquet) modes m , for which the term in the Hamiltonian with a phase factor $e^{\mp i\Omega t}$ induces a transition between the Floquet mode m and $m \pm 1$.

The matrix representation of Eq. (6) is in general infinite-dimensional, since m takes from $-\infty$ to $+\infty$. In the present case, however, $S_{\text{tot}}^z - m$ is a good quantum number because the term S_{tot}^{\pm} appears in the Hamiltonian with a phase factor $e^{\mp i\Omega t}$, so that the term simultaneously changes the Floquet index and S_{tot}^z by ± 1 , re-

spectively, as shown in Fig. 5(a). This implies that the Floquet matrix can be put into a block-diagonal form as shown in Fig. 5(b). We call the Hamiltonian that acts within the blocks an “irreducible Floquet Hamiltonian.” If the system size (i.e., the total number of spins) is finite, each block is finite-dimensional since S_{tot}^z is bounded as $-N \leq S_{\text{tot}}^z \leq N$. Thus, even in the presence of the time-dependent external field, we can readily solve the eigenvalue equation with an exact diagonalization as far as finite systems are concerned.

Interestingly, the irreducible Floquet Hamiltonian is formally equivalent to the Hamiltonian for an $S = 1$ chain with longitudinal and transverse magnetic fields. Since

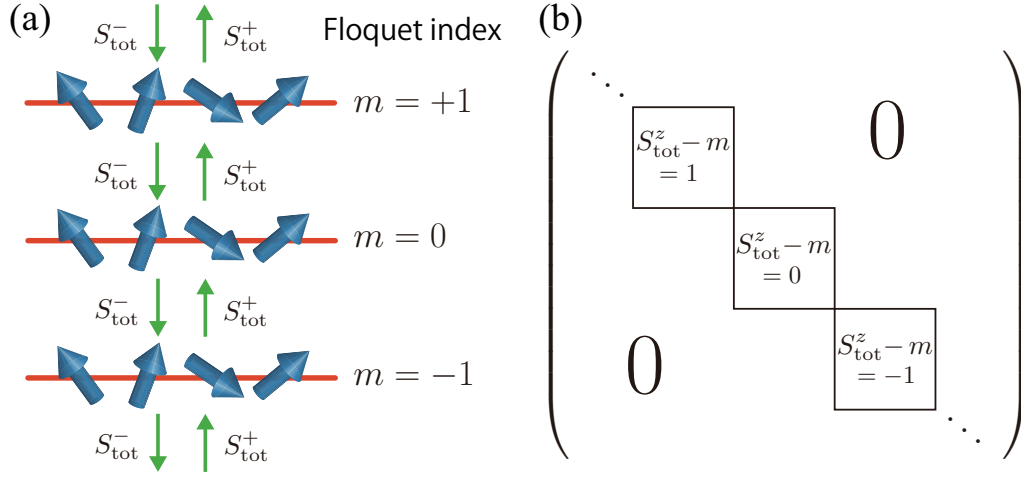


FIG. 5: (a) Structure of the Floquet Hamiltonian (7) for a spin system in a circularly-polarized light field. (b) Block-diagonal structure of the Floquet Hamiltonian reshuffled in terms of the good quantum number $S_{\text{tot}}^z - m$.

the S_{tot}^\pm term connects the sectors which have S_{tot}^z differing by ± 1 , $B(= 2A)$ acts as the transverse magnetic field. On the other hand, Ω act as the longitudinal magnetic field. We can see this because the matrix components in the same S_{tot}^z sector are $H_0 - m\Omega$, which translates to $H_0 - \Omega S_{\text{tot}}^z$ since $S_{\text{tot}}^z - m$ is constant within each irreducible Floquet matrix (where the constant, being irrelevant, can be set to 0). We end up with an effective Hamiltonian,

$$H_{\text{Ir.Fl.}} = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2 - \Omega S_{\text{tot}}^z - B S_{\text{tot}}^x. \quad (8)$$

The time evolution of M^z can then be calculated from the exact diagonalization result for the eigenvalues ($\{\epsilon_\alpha\}$) and eigenvectors ($\{|\Phi_\alpha\rangle\}$) of Eq. (8). We can reconstruct the solution of the original time-dependent

Schrödinger equation as $|\Psi(t)\rangle = \sum_\alpha c_\alpha |\Psi_\alpha(t)\rangle$, where $|\Psi_\alpha(t)\rangle \equiv \sum_m e^{-i(\epsilon_\alpha + m\Omega)t} |\Phi_\alpha^m\rangle$, and the coefficient c_α , with $\sum_\alpha |c_\alpha|^2 = 1$ for normalization, is determined from the initial condition. For example, in the case in which the application of laser begins suddenly at $t = 0$, $c_\alpha = \langle \Psi_\alpha(t=0) | \Psi_0 \rangle$, where $|\Psi_0\rangle$ is the initial state, i.e., the ground state (GS) of $\mathcal{H}_0 = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2$. The magnetization per spin M^z then evolves as

$$M^z(t) = \sum_{\alpha,m} |c_\alpha|^2 \langle \Phi_\alpha^m | S_{\text{tot}}^z / N | \Phi_\alpha^m \rangle + \sum_{\alpha < \beta, m} [c_\beta^* c_\alpha e^{i(\epsilon_\beta - \epsilon_\alpha)t} \langle \Phi_\beta^m | S_{\text{tot}}^z / N | \Phi_\alpha^m \rangle + \text{h.c.}],$$

where the first (second) term is the t -dependent (t -independent) part.